# Subsurface Science Needs for the Hanford Site

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## Background

The U.S. DOE Hanford Site in south central Washington state lies along the Columbia River and is one of DOE's largest legacy waste management sites. Enormous radionuclide inventories exist in below-ground storage in dewatered, single shell high level waste tanks in Hanford's twelve tank farms and in the subsurface where chemical and radioactive contaminants were discharged to the soil surface and to the deep vadose zone. Some of these discharges were from past leaks from the single-shell tanks and others were from waste disposal to engineered structures including cribs, trenches, French drains, infiltration ponds, and solid waste burial sites. Numerous contaminant plumes exist in the vadose zone (e.g., containing U, $^{99}Tc$, $^{90}Sr$, $^{137}Cs$, $CCl_4$, Cr(VI)) and the unconfined aquifer system (e.g. $^{3}H$, $^{129}I$, $^{99}Tc$, $^{90}Sr$, U, Cr(VI), $CCl_4$) that underlies the site at depths of 5-95 m. A small number of these groundwater plumes discharge contaminants to the Columbia River. Chemical and radioactive contaminants with long half-life, significant environmental mobility (e.g. over potential kilometer distances in groundwater), and potential health/ecological effects (chemical or radiologic) [e.g., U, $^{99}Tc$, $^{129}I$, $CCl_4$, Cr(VI)] drive risk at Hanford and are of major, long-term concern.

## Site Status

The Hanford Site has been under active remediation for over ten years and significant progress has been made in understanding the subsurface contamination problem and in remediating the Site to prevent the further spread of contaminants to the Columbia River. Massive amounts of contaminated soil has been excavated and moved from the Columbia River corridor and stabilized in a regulated landfill in the center of the site. Spent fuel stored in water in the K-Basins near the Columbia River was successfully retrieved, moved and stabilized. Vadose zone contaminant plumes beneath leaked high level waste tanks of various composition (e.g., SX-108, BX-102, TX-104, T-106), and high risk crib sites (e.g., Z-9) and trenches (e.g. B-26) have been rigorously characterized and studied by Hanford and EMSP scientists providing valuable insights on geochemical and hydrologic processes controlling contaminant migration. Defensible corrective measures have been proposed based on this science. Pump and treat operations have been conducted in multiple locations and found to be effective for $^{99}Tc$, Cr(VI), and $CCl_4$ to certain degree, but ineffective for more strongly-sorbing U and $^{90}Sr$. A permeable, reducing barrier was emplaced along the Columbia River to mitigate the discharge of groundwater with Cr(VI) to the river. That barrier is generally effective, but improvements are needed. The site-wide groundwater and river monitoring program has continued, cooperating with the activities noted above providing comprehensive temporal information on contaminant plume movement and impact on river water quality. An extensive information base (e.g., reports, publications, and monitoring data) on all of these activities and more can be found at: www.hanford.gov and at Groundwater Remediation Project website: http://www.hanford.gov/cp/gpp/.

**<u>Role of Science</u>**

Research performed by the EMSP investigators in collaboration with site-funded activities has played an important recent role at the Hanford Site. Close partnership between the EMSP Program and Hanford began in 1997 with an EMSP call focused on Hanford vadose zone issues. That research resolved key issues related to subsurface water and contaminant migration in the vadose zone which provided crucial technical support for regulatory decisions on corrective actions for the tank farms. The EMSP vadose zone research campaign and more recent research have yielded many important conclusions, as exemplified by the following:

- Extremely high Na concentrations and elevated waste temperature expedited Cs subsurface migration in the SX tank farm.
- The base (waste)-induced hydrolysis of ferrous silicates in vadose zone sediments promoted the reduction and unexpectedly high retardation of Cr(VI) in the SX tank farm.
- Repetitive, centimeter scale silt lenses that are common in Hanford vadose zone sediments promote significant lateral migration of waste fluids and greatly enhance physical retardation and travel times to groundwater.
- One of the largest, single point releases of U at Hanford has been immobilized approximately 30 m below the surface by precipitation of uranyl-silicates in intergrain fractures of feldspar-containing lithic fragments.
- The 300 A U(VI) groundwater plume has dissipated more slowly than expected after source term removal because low concentration vadose zone residuals continue to dissolve/desorb to porewater and the sorption complexes responsible for retardation in both the vadose zone and groundwater exhibit slow kinetics.

The results of these research activities are listed in the attached bibliography of scientific publications.

Ongoing subsurface EMSP research at Hanford seeks to understand U geochemistry in the vadose zone and the unconsolidated aquifer, factors controlling intergrain immobilization of $CCl_4$, and the role of centimeter scale heterogeneities on moisture flow vectors and the physical retardation of $^{99}Tc$ in the vadose zone, among other issues.

**<u>Current Scientific Needs</u>**

In spite of significant progress made by EMSP researchers, Hanford Site investigators, and others in understanding subsurface geochemical and hydrologic processes regulating contaminant migration and fate, significant subsurface science needs remain. Past scientific research has resolved many critical issues at the Hanford Site, others have been investigated but remain unresolved, and anomalous observations at sites recently characterized and evolving plans for remediationg waste sites pose new vexing challenges and questions. The following represent important current needs and their justification.

1.) Rigorous conceptual and numeric models along with appropriate thermodynamic and kinetic parameters are needed that describe the geochemical interactions and reactive transport behavior of U(VI) and $^{129}I$ in the varied geohydrochemical environments and sediment facies found in the Hanford vadose zone and unconfined aquifer. The influence of waste source composition, temperature, and other properties on reaction chemistry requires explicit consideration. Uranium transport has largely been resolved for the BX-102 overfill at the B-BX-BY Tank Farm, but questions remain regarding uranium transport at other tank farms and near the Columbia River at the 300 Area.

2.) An improved hydrochemical conceptual model is needed for the subsurface behavior of $CCl_4$ at the Hanford site including its migration behavior as an organic fluid through the water-unsaturated vadose zone and its reaction chemistry as a dissolved solute in both the vadose zone and groundwater. Information is critically needed on the sorption and degradation chemistry of $CCl_4$ in Hanford and Ringold formation lithofacies that differ in texture and mineralogy. Remedial strategies that are consistent with the hydrochemical model are also sought for dissolved $CCl_4$ in deep Hanford groundwaters (e.g., > 35 m below ground surface).

3.) Effective, cost-feasible remedial methods are sought that are consistent with known hydrologic and geochemical models of the Hanford vadose zone and unconfined aquifer to prevent: i.) further downward migration of deeply distributed U(VI) and $^{99}Tc$ in the vadose zone and ii.) lateral migration of U(VI) in Hanford's unconfined aquifer and its discharge to the Columbia River.

4.) In support of eventual Site closure, research is needed into water and vapor flow through Hanford sediments at low moisture contents (<10%). Low native infiltration rates will be further reduced as surface barriers are placed over waste sites to prevent or slow migration of in-ground contaminants. Flow and transport of water and contaminants at low moisture contents needs to be investigated in the context of fine-textured layers present at the Hanford Site. These fine-textured layers have been shown to exert a major influence on water and contaminant migration in Hanford's vadose zone.

5.) Reactive transport modeling that spans multiple spatial and temporal scales is needed for representative Hanford subsurface hydrogeologic environments and key waste sites (vadose zone and groundwater plumes) to document the credibility of posed conceptual models of reaction and transport, to evaluate the influences of centimeter to meter scale heterogeneities in physical and chemical properties, to assess the implications of process coupling, and to provide improved and scientifically credible projections of future moisture and solute transport. Investigators are encouraged to access extensive data bases on experimental and contaminated field sites at Hanford, and to pose additional research as required to enable defensible, state-of-science simulations of high scientific and site-specific value.

6.) In support of long-term Site stewardship, research is needed on long-term, in-ground geochemical and/or biogeochemical processes (and their rates and products) that may immobilize, mobilize, or otherwise change the behavior of in-ground contaminants including U, $^{129}I$, $^{99}Tc$, Pu/Am, Cr(VI) and $CCl_4$. Investigations using representative Hanford sediment facies and contaminant matricies poised over appropriate ranges in water potential are essential.

7.) On a fundamental level, robust methods are needed for describing and modeling water and reactive solute behavior in detail at the pore scale and the derivation of constitutive properties from these activities that can be used at larger scales. Beneficial research would evaluate relationships between pore morphology, topology, connectivity, and reaction site distribution with scale-dependent properties such as permeability, diffusivity, dispersivity, and reactivity.

8.) The Columbia River is the primary exposure vector by which Hanford contaminants contact higher trophic organisms and man (today but not in the past). Hanford contaminants enter the Columbia River through the groundwater-river interface that includes the hyporeic zone in the riverbed. The chemistry, microbiology, and hydrology of groundwater-river interface, and the hyporeic zone explicitly, have been monitored and investigated to limited extent, but additional research is required to understand the influence of river stage on contaminant concentrations, contaminant biotransformations and biouptake, and the integrated processes that regulate contaminant flux from groundwater to the Columbia River.

# Journal Publications Related to the Hanford Site


**<u>2004</u>**

Ainsworth, C. C., J. M. Zachara, K. Wagnon, S. McKinley, S. C. Smith and P. L. Gassman. 2004. Impact of highly basic solutions on Hanford subsurface sediments and its sorption of Cs. *Geochimica et Cosmochimica Acta* (Submitted).

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